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Numerical modelling and laboratory studies on the removal of Direct Red 23 and Direct Red 80 dyes from textile effluents using orange peel, a low-cost adsorbent

F. Doulati Ardejani [a,b,*], Kh. Badii [b], N. Yousefi Limaee [b], N.M. Mahmoodi [b], M. Arami [b,c], S.Z. Shafaei [a,b], A.R. Mirhabibi [b,d]

[a] *Faculty of Mining and Geophysics, Shahrood University of Technology, Shahrood, Iran*
[b] *Environmental Science and Engineering Department, Iran Colour Research Centre, Tehran, Iran*
[c] *Textile Engineering Department, Amir Kabir University of Technology, Tehran, Iran*
[d] *Department of Metallurgy, University of Science and Technology of Iran, Tehran, Iran*

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Abstract

Wastewater containing direct dyes discharged from various industries, in particular, textile industry often causes many environmental problems. Considering the possibility of the adsorption process of synthetic dyes and their removal from aqueous solutions using orange peel as an eco-friendly and low-cost adsorbent before discharging to the receiving environment is the most important task. This paper presents a numerical finite element model to simulate the removal of dissolved textile synthetic dyes from wastewater taking into consideration both linear and the Langmuir isotherms to describe adsorption process. The CTRN/W model was integrated with SEEP/W model and modified to solve mathematical equations describing the problem at hand. The modelling accuracy was first verified with an analytical equation for a reactive mass transport problem including advection, dispersion and linear adsorption processes through groundwater flow medium. The modelling results were then compared to those results obtained from experimental tests for the removal of Direct Red 23 and Direct Red 80 dyes from textile wastewater using orange peel adsorbent. The model predictions somewhat agree with the experimental measurements. It was found that the adsorption process by orange peel adsorbent could be well described by the Langmuir isotherm. Furthermore, a pseudo-second order kinetics showed to be well-suited with the rate of sorption. Although the generation of such wastewater by the different industries is nearly unavoidable, the results of model presented here can help to design an appropriate environmental management strategy to minimise the adverse impacts caused by industrial wastewater.

Keywords: Numerical modelling; Finite element; Direct Red 23 and 80; Adsorption; Isotherm; SEEP/W; CTRN/W

1. Introduction

Textile industries produce huge amounts of polluted effluents that are normally discharged to surface water bodies and groundwater aquifers. These wastewaters cause many damages to the ecological system of the receiving surface water [1,2] and create a lot of disturbance to the groundwater resources.

Most dyes used in textile industries are stable to light and are not biologically degradable [3]. Furthermore, they are resistant to aerobic digestion [4]. In order to reduce the risk of environmental pollution from such wastes, it is necessary to accurately treat them before discharging to the receiving environments.

Considerable efforts have been made by many researchers to find appropriate treatment systems in order to remove pollutants and impurities of wastewaters emanated from different

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* Corresponding author. Faculty of Mining and Geophysics, Shahrood University, Shahrood, Iran. Tel.: +98 273 3344004; fax: +98 273 3335509.
E-mail address: fdoulati@shahrood.ac.ir (F. Doulati Ardejani).

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industries, in particular, textile industry [1–7]. Many chemical and physical methods of dye-removal, including photocatalytic degradation [7], membranes [8] and adsorption techniques [3,4] have been used from time to time. Adsorption process is noted to be superior to other removal techniques because it is economically cost effective, simple [2] and it is capable to efficiently treat dyes in more concentrated form [3].

Although many experimental works have been conducted to assess the capability and the performance of various adsorbents for the removal of dyes from the textile industry, little research has been done to model dye-removal process from the textile wastewaters and to evaluate the significance of the effect of major parameters on the percent of dye adsorption. Numerical models are valuable tools to provide insight into the adsorption process. A numerical model enables the prediction of future events. It may indicate which factors in a real system are most important from a sensitivity point of view. A model can help to design, optimise and predict the performance of field remediation and polluted waters treatment programs.

A numerical finite element model using CTRN/W software has been presented to simulate removal of dissolved textile dyes from wastewaters taking into account that adsorption is the main mechanism for the removal process. The adsorption efficiency of two different dyes was considered in the model. Furthermore, the effects of the initial concentrations of both dyes on the quantity of dye removal were evaluated. The results obtained from the model presented here can help to design an appropriate wastewater management strategy to minimise the socio-economic and environmental impacts of textile effluents.

A commercial numerical finite element software called CTRN/W [9] was modified to simulate the removal of direct dyes from wastewater. This software can be used to model the transport of contaminants through porous media such as soil and rock. The main processes incorporated in CTRN/W are diffusion, dispersion, adsorption, radioactive decay and density dependencies.

CTRN/W is coupled with SEEP/W, another finite element software [9] that creates the finite element grid and calculates the flow velocity for a transport problem in which the movement of the contaminants is also considered. CTRN/W utilises the SEEP/W finite element grid to model adsorption process of the direct textile industry wastewater. In all CTRN/W simulation, the transport equations solved by CTRN/W are expressed as:

$$\left(\theta + \rho_d \frac{\partial S}{\partial C}\right)\frac{\partial C}{\partial t} = \theta D \frac{\partial^2 C}{\partial x_j^2} - U_j \frac{\partial C}{\partial x_j} - KS\rho_d - K\theta C \qquad (1)$$

where

θ = volumetric water content (dimensionless);
C = concentration (mg/l);
D = hydrodynamic dispersion coefficient (mm^2/s);
x_j = Cartesian coordinates (mm);
U_j = Darcian velocity in the x_j direction (mm/s);
ρ_d = bulk density of the medium (1/1000 mg/mm^3);
S = concentration in the solid phase (mg/g);
t = time (s);
K = decay constant (1/s).

The decay constant K can be related to the half-time T (s) for decaying substance as follows [10]:

$$T = \frac{0.693}{K} \qquad (2)$$

Eq. (1) was modified to model the removal of dissolved textile synthetic dyes from wastewater by assigning appropriate initial and boundary conditions to the model.

2. Experimental

To consider the capability of the numerical model for the simulation of the removal of pollutants from the industrial effluents in order to reduce environmental pollution risk, the results obtained from an experimental test were selected for the comparison purposes. In this laboratory test, the potential for the removal of Direct Red 23 and Direct Red 80 dyes from textile effluents by sorption process using orange peel adsorbent was investigated.

2.1. Sorbent preparation

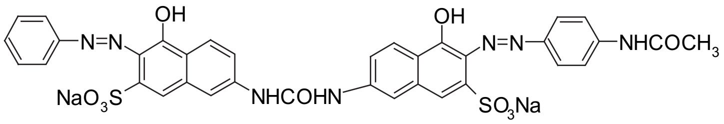
The orange peel was obtained from a local fruit field in the north of Iran. The orange peels were first washed to remove any adhering dirt and then were dried, crushed and sieved. They were air dried at room temperature for 36 h. After drying, the peels were passed through a 3.36 mm sieve.

2.2. Reagents and solutions

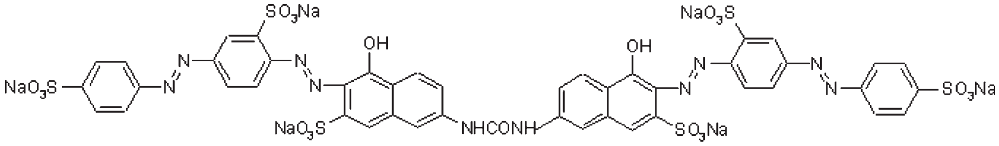
Direct Red 23 and Direct Red 80 were obtained from Ciba Ltd. and were used without any further purification. The chemical structures of these dyes are shown in Fig. 1. Distilled water was throughout employed as solvent. Working solutions of 50 and 75 mg/l were prepared for both dyes. The pH adjustments of the solutions were made by adding a small amount of 2 M H_2SO_4 or NaOH.

2.3. Adsorption procedure

The adsorption measurements were conducted by mixing various amounts of orange peel (0.5–2 g) for Direct Red 23 and (0.2–1 g) for Direct Red 80 in jars containing 250 ml of a dye solution (50 mg/l) at pH ranging from 2 to 10. pH studies were performed to determine the optimum pH at which maximum colour removal could be achieved with the adsorbent for each dye. Although not given here, the experiments were carried out at various concentrations of 25, 50, 75, 100, and 125 mg/l using 2 g adsorbent for Direct Red 23 and 1 g adsorbent for Direct Red 80 at pH 2 and temperature 25 °C for 24 h to attain equilibrium conditions [11]. An FC6S-VELP (Scientifica) jar test was used for agitating purpose. Different agitation rates ranging from 30 to 200 rpm had no



Direct Red 23



Direct Red 80

Fig. 1. Chemical structures of the two dyes used in this study.

significant change on adsorption process. The changes of absorbance were determined at certain time intervals (15, 30, 45, 60, 120, 180, 240, 300, 360, 420, 480, and 1440 min) during the adsorption process. The absorbance measurements were made using a CECIL 2021 spectrophotometer. After experiments, the sorbent was separated from the solution by Hettich EBA20 centrifuge and dye concentration was then determined. The results were fitted to the Langmuir non-linear adsorption isotherm with correlation coefficients of 0.9762 and 0.9997 for Direct Red 23 and Direct Red 80, respectively (see Table 1).

3. Results and discussion

3.1. Adsorption isotherms

Reactions between solutes and the surfaces of solids play a crucial role in controlling the chemistry of industrial effluents. Sorption isotherms are often used to describe interactions between solutes and solid matrix [12]. The adsorption isotherms illustrate the relationships between equilibrium concentrations of adsorbate in the solid phase (S), and in the liquid phase (C) at constant temperature [5]. The distribution of pollutant such as dye between the adsorbent and the dye solution under equilibrium conditions is important in understanding the capacity of the adsorbent for the dye removal [4]. Sorption isotherms are obtained in the laboratory using batch tests [10,13]. Two of the most common non-linear isotherm models are the Langmuir and the Freundlich isotherms [5,10,12–14]. Langmuir isotherm is:

$$S = \frac{Q_0 K_L C}{1 + K_L C} \quad (3)$$

The general form of the Freundlich isotherm is:

$$S = K_F C^n \quad (4)$$

where

S = quantity of mass sorbed on the solid surface (mg/g);
C = equilibrium concentration of the solution (mg/l);
Q_0 = maximum adsorption capacity;
K_L = Langmuir constant;
K_F = partition coefficient indicating adsorption capacity;
n = Freundlich exponent, generally ranging between 0.7 and 1.2.

Table 1
Linearised isotherm coefficients for dyes

Dye	Q_0	K_L	R^2	K_F	n	R^2
Direct Red 23	10.718	0.308	0.9762	4.431	4.189	0.8896
Direct Red 80	21.052	0.429	0.9997	9.384	4.160	0.9549

A special case of the Freundlich isotherm with $n = 1$ is called linear isotherm as given by the following equation:

$$S = K_d C \quad (5)$$

where

K_d = slope of the linear sorption isotherm, also recognised as the distribution coefficient (ml/g).

The distribution coefficient K_d can be related to the retardation factor R which describes the effects of the linear adsorption [15]:

$$R = 1 + \frac{\rho_d}{\varphi} K_d \quad (6)$$

where

ρ_d = dry density of the adsorbent;
φ = porosity.

The equilibrium isotherms for the adsorption of the two dyes Direct Red 23 and Direct Red 80 on orange peel determined for the initial concentration of 50 mg/l are shown in Figs. 2 and 3, respectively. These non-linear isotherms were obtained in a constant temperature. The empirical parameters of the Langmuir and the Freundlich isotherms are given in Table 1. The parameter R^2 in Table 1 is the squared correlation coefficient.

3.2. Adsorption kinetics

The study of the adsorption kinetics is a main factor for designing an appropriate adsorption system. The adsorption of dyes from aqueous phase to solid phase can be well described

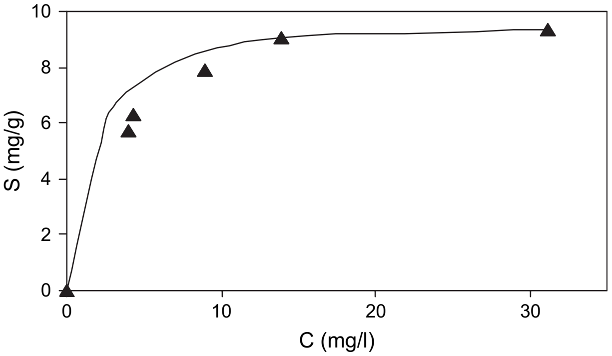


Fig. 2. Experimental (▲) and predicted (—) isotherms for adsorption of Direct Red 23 dye.

as a reversible reaction under an equilibrium condition established between two phases [4]. In order to consider the kinetics effects, the following Lagergren pseudo-first order equation can be used to determine the rate constants.

$$\log(q_e - q_t) = \log q_e - \frac{K_{1,\mathrm{ad}}}{2.303}(t) \tag{7}$$

where

q_e = quantity of dye adsorbed at equilibrium (mg/g);
q_t = quantity of dye adsorbed at time t (mg/g);
$K_{1,\mathrm{ad}}$ = pseudo-first order rate constant (min^{-1});
t = time (min).

In many cases Eq. (7) cannot fully describe the kinetics of the adsorption process. In such cases, a pseudo-second order expression may be used. This model is:

$$\frac{t}{q_t} = \frac{1}{K_{2,\mathrm{ad}} q_e^2} + \frac{1}{q_e}(t) \tag{8}$$

where
$K_{2,\mathrm{ad}}$ = pseudo-second order rate constant ($\mathrm{g\,mg^{-1}\,min^{-1}}$).

In this study, both first and second order kinetics model were considered for the adsorption process. The kinetics parameters are summarised in Table 2. As shown in Table 2, the adsorption kinetics of Direct Red 23 and Direct Red 80 dyes follows the pseudo-second order rate expression with squared correlation coefficient greater than 0.99.

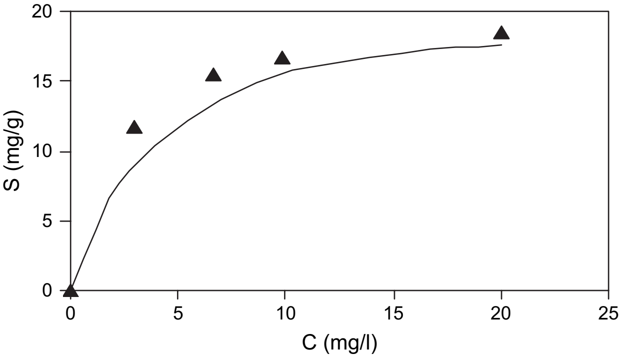


Fig. 3. Experimental (▲) and predicted (—) isotherms for adsorption of Direct Red 80 dye.

Table 2
Kinetics parameters for the pseudo-first and -second order models

Dye	Concentration (mg/l)	$K_{1,\mathrm{ad}}$	R^2	$K_{2,\mathrm{ad}}$	R^2
Direct Red 23	50	0.103	0.5827	0.374	0.9980
Direct Red 80	50	0.066	0.2854	0.201	0.9996
Direct Red 23	75	0.126	0.9841	0.307	0.9996
Direct Red 80	75	0.0461	0.9664	0.0603	0.9947

3.3. Modelling

3.3.1. Model validation

The present model was checked by comparison with analytical solutions for one-dimensional advection–dispersion contaminant transport equation with linear adsorption process and under steady-state water flow condition. This example was mostly taken from the CTRN/W documentation [9] and slight modifications were made. In this case, the volumetric water content was 0.5. The bulk density of the medium was 0.5 $\mathrm{g/mm^3}$. The dispersivity (α_L) was 150 mm, and the molecular diffusion coefficient $\left(D_{\mathrm{dif}}\right)$ was negligible. The Darcian velocity was equal to 0.15 mm/s. The hydrodynamic dispersion coefficient was calculated as:

$$D = D_{\mathrm{dif}} + \alpha_L U/\theta = 0 + 150 \times 0.15/0.5 = 45\ \mathrm{mm^2/s}$$

A retardation factor of 2 was considered for this example. The following equation [13] was first used to solve the problem analytically.

$$C(x,t) = \frac{C_0}{2}\left\{\mathrm{erfc}\left(\frac{Rx - vt}{2\sqrt{RDt}}\right) + \exp\left(\frac{vx}{D}\right)\mathrm{erfc}\left(\frac{Rx + vt}{2\sqrt{RDt}}\right)\right\} \tag{9}$$

To solve the problem numerically, a 3500 mm horizontal profile was divided into 50 non-uniform quadratic elements using SEEP/W model. The model contained 253 nodes and 60 mm thickness (Fig. 4).

A constant head of 2500 m was assigned at the inlet boundary of the model. A constant head of 2490 m was also maintained at the outer boundary. The permeability was considered $5.2542 \times 10^{+1}$ mm/s. These settings produced a constant Darcian velocity equal to 0.15 mm/s. The volumetric water content was set to 0.5. A simulation was then performed at a steady-state condition. The model for this part of the simulation was named *TRANSPORT.SEZ*.

To model the main part of the simulation by the CTRN/W software, the *TRANSPORT.SEZ* model constructed by the SEEP/W software was first imported and it was named *TRANSPORT.CTZ*. A first-type or Dirichlet boundary condition was considered at inlet ($x = 0$), with $C(0,t) = 1\ \mathrm{g/mm^3}$.

An initial value of 0 was maintained at $t = 0$. A value of 0 was specified at outlet boundary ($C(\infty,t) = 0$). Fifty time steps were considered. A total iteration of 10 was assigned for the simulation. A central difference time integration scheme was selected. The longitudinal dispersivity was set

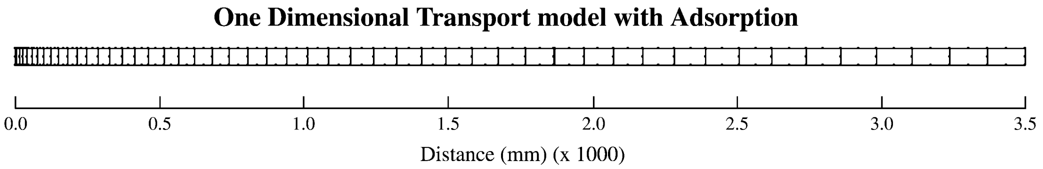


Fig. 4. Finite element representation of the reactive transport model.

to 150 mm. The bulk density was defined as a constant value 0.5. In order to compare CTRN/W predictions with those results obtained from the analytical solution (Eq. (9)) when R is equal to 2, the equivalent retardation factor R_e in the CTRN/W formulation, must be set to 1.

The readers are referred to the CTRN/W documentations [9] for more details. To set R_e, an adsorption function was defined as a straight line with a slope of 1.

The comparison of the relative concentrations predicted by the numerical model (line) and those calculated using Eq. (9) (triangles) are shown in Fig. 5. The agreements between two methods in the case where the adsorption process is incorporated are quite close.

As the figure shows, the results were given for different elapsed times of 3000, 4000 and 5000 s with adsorption (continuous lines) and without adsorption (broken lines). Adsorption in the transport process retards the contaminant migration process.

3.3.2. Modelling predictions for dye removal

After verification, the model was then run to perform simulation of dye removal from textile wastes, taking into consideration that the adsorption is the main mechanism for dye removal. The simulation was carried out in three different runs as given below.

Run 1: The first step in a CTRN/W simulation is to establish a finite element mesh using SEEP/W model. A one-dimensional finite element mesh consisting of 20 elements and 103 nodes was therefore constructed using SEEP/W. The hydraulic head differential and hydraulic conductivity were selected to produce no velocity. The volumetric water content θ was defined as a constant 0.5. A steady-state simulation was then performed.

Run 2: The associated SEEP/W file was included with the CTRN/W software. To specify the input data, the material properties box was selected from the KeyIn menu. The longitudinal and transverse dispersivities were set to 1.0×10^{-20} mm. A value equal to the initial dye concentration in the textile waste was specified at all nodal points of the finite element mesh. A small time step of 1.0×10^{-20} s was assigned to consider the simulation as a steady-state condition. The SOLVE window was then used to perform this steady-state simulation and provide the initial conditions for the main transient modelling of the dye removal process. This file should be used as an initial condition for the transient simulation of the dye removal process.

Run 3: To perform the final stage of the simulation, the dye initial concentration was first removed from all nodal points. Fifteen time steps were assigned for the simulation. Results are presented for time steps 6–15 with total elapsed times of 180, 360, 540, 720, 900, 1200, and 1500 s. A total iteration of 100 was assigned for the simulation. A central difference time integration scheme was considered. In the case of a linear adsorption, Eq. (1) reduces to:

$$R\frac{\partial C}{\partial t} = -KS\rho_d \tag{10}$$

Taking into consideration Eq. (5), Eq. (10) can then be written as:

$$R\frac{\partial C}{\partial t} = -KK_dC\rho_d \tag{11}$$

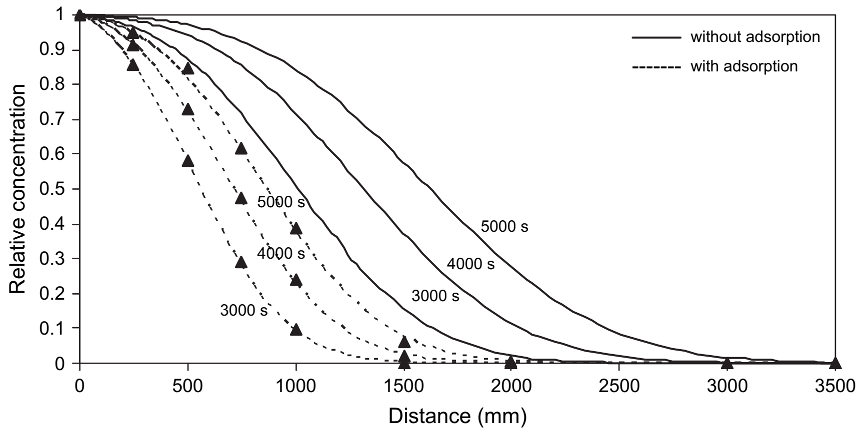


Fig. 5. Comparison of analytical (▲) and numerical (—) solutions for mass transport problem with linear adsorption through groundwater system.

Table 3
Parameters used for simulation of linear adsorption case

Input parameter	Dye type			
	Direct Red 23 initial concentration (mg/l)		Direct Red 80 initial concentration (mg/l)	
	50	75	50	75
Longitudinal dispersivity (m)	1×10^{-20}	1×10^{-20}	1×10^{-20}	1×10^{-20}
Transverse dispersivity (m)	1×10^{-20}	1×10^{-20}	1×10^{-20}	1×10^{-20}
Decay half-time (s)	68.88	60.083	48.39	600
Dry density (1/1000 mg/mm^3)	1×10^{-20}	1×10^{-20}	1×10^{-20}	1×10^{-20}
Retardation factor	0.4035×10^{20}	0.5635×10^{20}	1.05×10^{20}	0.3202×10^{20}
Distribution coefficient	1.5	2.095	3.973	1.0674

Table 4
Model input parameters used for non-linear simulation

Input parameter	Dye type			
	Direct Red 23 initial concentration (mg/l)		Direct Red 80 initial concentration (mg/l)	
	50	75	50	75
Longitudinal dispersivity (m)	1×10^{-20}	1×10^{-20}	1×10^{-20}	1×10^{-20}
Transverse dispersivity (m)	1×10^{-20}	1×10^{-20}	1×10^{-20}	1×10^{-20}
Decay half-time (s)	112	111.18	97	594
Molecular diffusion (m^2/s)	1×10^{-10}	1×10^{-10}	1×10^{-10}	1×10^{-10}

where

K = adsorption kinetics.
Eq. (11) yields:

$$R\frac{\partial C}{\partial t} = -K_1 C \tag{12}$$

K_1 may be calculated in terms of adsorption kinetics, distribution coefficient and bulk density as given below:

$$K_1 = KK_d\rho_d \tag{13}$$

It should be noted that CTRN/W multiplies the volumetric water content, θ in the sink term in Eq. (12). To take it into consideration, it is therefore necessary to modify Eq. (13) by dividing it by θ. Hence, Eq. (13) can now be rewritten as:

$$K_1 = KK_d\rho_d/\theta \tag{14}$$

K_1 can be related to the half-time T using Eq. (2). The settings made for a linear adsorption case are given in Table 3.

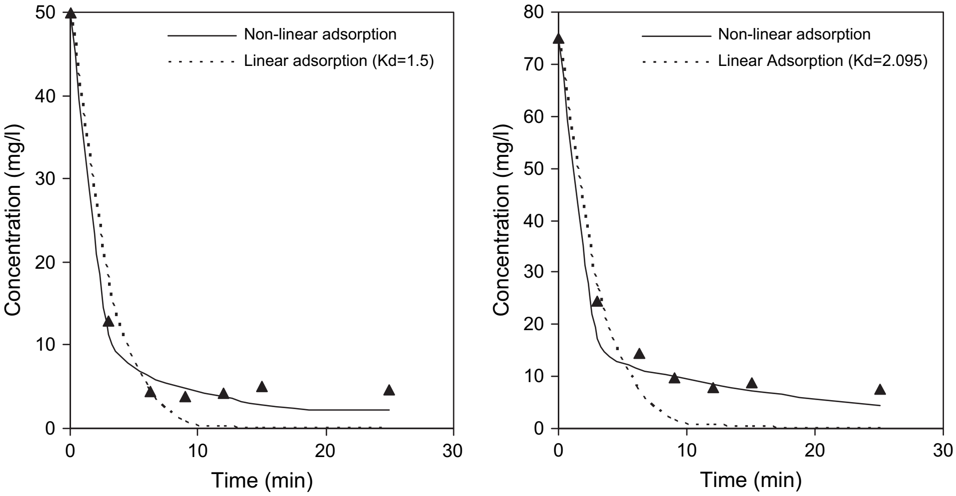
As Table 3 shows, the dry density ρ_d was set to 1×10^{-20}. This removes the last term $(-K\theta C)$ in Eq. (1). Hence, the following slight modification is also necessary to calculate the equivalent retardation factor R_e:

$$R_e = \theta + (0.538\rho_d K_d)/1 \times 10^{-20} \tag{15}$$

where the value of 0.538 is the actual amount of the dry density in terms of g/cm^3. For a non-linear adsorption process, the model input data are listed in Table 4.

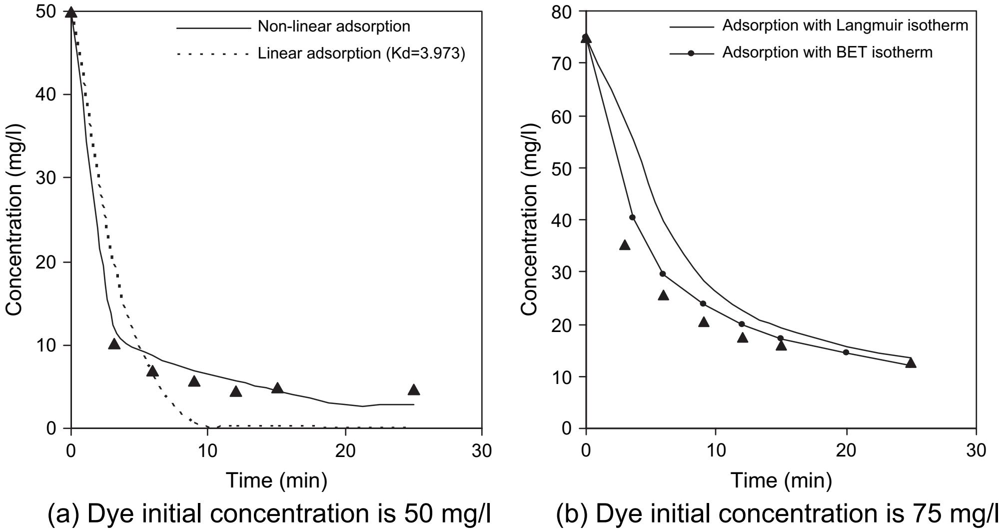
3.3.3. Comparison of modelling and experimental results

Figs. 6 and 7 show a comparison between the numerical model shown as solid lines and the measured data shown as triangles for the rate of dyes sorption by the orange peel and the agreement was somewhat close. The results indicated that the rates of removal of the direct dyes are sensitive to their



(a) Dye initial concentration is 50 mg/l (b) Dye initial concentration is 75 mg/l

Fig. 6. Comparison of measured (▲) and predicted (—) rate of sorption of dye Direct Red 23 on orange peel at pH 2 and initial concentrations of 50 and 75 mg/l.



(a) Dye initial concentration is 50 mg/l (b) Dye initial concentration is 75 mg/l

Fig. 7. Comparison of measured (▲) and predicted (—) rate of sorption of dye Direct Red 80 on orange peel at pH 2 and initial concentrations of 50 and 75 mg/l.

initial concentrations. When the initial concentrations of both Direct Red 23 and Direct Red 80 dyes were increased from 50 to 75 mg/l, the rate of removal by the orange peel adsorbent decreased. In these figures, the effect of the linear adsorption model (broken line) is also considered. As illustrated, the linear adsorption model could not describe the dye removal rate while the removal of dyes from textile effluents may be well described by the non-linear adsorption process.

As Fig. 7(b) shows, differences between the predicted values and measured quantities are high if the model considers the Langmuir isotherm. While a close agreement was achieved between the simulated results and measured values when a Brunauer–Emmett–Teller (BET) model [16] was taken into account as the adsorption isotherm.

4. Conclusion

A numerical finite element model has been developed to simulate the removal of the direct dyes from textile effluents taking into consideration both linear and the Langmuir isotherms to describe adsorption process. To solve the problem numerically, the CTRN/W software was coupled with the SEEP/W model. The parameter values in the model were obtained from an experimental test for the removal of dyes Direct Red 23 and Direct Red 80 from textile wastewaters using orange peel as a low-cost adsorbent. The results of the model somewhat agreed with the measured quantities. It was found that the adsorption process by orange peel adsorbent could be well described by the Langmuir isotherm. Furthermore, a pseudo-second order kinetics showed to be well-suited with the rate of sorption. Moreover, the rate of dye removal from the textile effluents is sensitive to the initial concentration of dye in solution phase. When the initial concentrations of both Direct Red 23 and Direct Red 80 dyes were increased from 50 to 75 mg/l, the rate of removal by the orange peel adsorbent decreased. Although the generation of such effluents by textile industry is almost unavoidable, the results of such numerical models can help to design an appropriate environmental management strategy to minimise the adverse impacts caused by industrial wastewaters.

Acknowledgments

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